High Pressure Study of Structural Phase Transitions and Superconductivity in $La_{1.48}Nd_{0.4}Sr_{0.12}CuO_4$

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We have determined the crystal structures and superconducting transition temperatures of $La_{1.48}Nd_{0.4}Sr_{0.12}CuO_4$ under nearly hydrostatic pressures in diamond anvil cells to 5.0 GPa and 19.0 GPa, respectively. Synchrotron x-ray powder diffraction measurements were used to establish the material's pressure-temperature structural phase diagram. Under pressure the superconducting transition temperature (T_c) increases rapidly from $T_c \approx 3$ K to a maximum value of 22 K at 5 GPa, a pressure slightly greater than required to stabilize the undistorted I4/mmm structure in the superconducting state.

Superconductivity is among the most remarkable physical phenomena discovered during the 20th century. Although the famous Bardeen-Cooper-Schrieffer theory of superconductivity can explain most experimental observations for conventional superconductors, the high-temperature superconducting cuprates have yet to be completely understood in a similar way.

Perhaps the simplest superconducting cuprates are based upon the parent compound lanthanum cuprate (La_2CuO_4), whose structure is shown in **Figure 1**. This material, when doped with increasing amounts of alkaline-earth ions such as Sr^{2+} (i.e. $La_{2-x}Sr_xCuO_4$), transforms from a Mott insulator to a metal, and then superconducts with a critical temperature (T_c) as high as 38 K when x = 0.15. Lanthanum cuprates for which some of the La^{3+} is replaced with both Sr^{2+} *and* Nd^{3+} (i.e. $La_{2-x-y}Nd_ySr_xCuO_4$) exhibit structural phase transitions involving tilts of the CuO_6 octahedra (**Figure 1**) that strongly suppress T_c. The suppression of T_c is most pronounced when the Sr^{2+} doping, x, is 1/8. These structural phase transitions are sensitive to external parameters such as pressure and temperature, and thus it is interesting to observe the effect of applied pressure at various temperatures upon the structure (and superconductivity) of these materials. Here we describe a set of x-ray powder diffraction experiments conducted for that purpose at beamline X7A.

The x-ray powder sample of composition $La_{1.48}Nd_{0.4}Sr_{0.12}CuO_4$ was immersed in a 4:1 ethanol:methanol mixture within a Merrill-Bassett diamond anvil cell that was then mounted in a displex closed-cycle refrigerator. A position-sensitive detector was used to detect the scattered x-rays of wavelength near 0.7 Å (energy of about 17 keV). Pressures were measured using laser-excited fluorescence from small ruby chips placed inside the diamond anvil cells, as well as by measurements of the lattice parameters of small quantities of NaCl or CaF_2 also included in the cells.

In **Figure 2** we show two typical x-ray diffraction patterns, obtained at pressures of 2.2 and 4.2 GPa, for a sample of composition $La_{1.48}Nd_{0.4}Sr_{0.12}CuO_4$. In **Figure 3** we show the structural phase diagram for this material (based upon the x-ray data), compared with the superconducting transition temperature, as a function of pressure. At ambient pressure this material has a low superconducting $T_c \approx 3$ K associated with the low-temperature tetragonal (LTT) structure and the special Sr^{2+} concentration of x = 1/8. Under pressure the LTT phase is first

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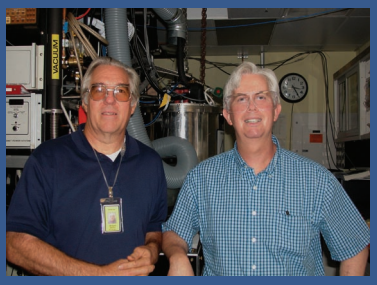

Authors (from left) Richard Harlow and Michael Crawford

replaced by the low-temperature orthorhombic LTO2 phase, then at pressures above 4.0 GPa the high-temperature tetragonal (HTT) structure remains stable to T = 10 K. The superconducting transition temperature increases dramatically under pressure and reaches a maximum value of T_c = 22 K near 5.0 GPa, coincident with the appearance of the undistorted HTT phase.

The results we describe here have implications for the presence of charge ordering in cuprates. The LTT structure of $La_{1.48}Nd_{0.4}Sr_{0.12}CuO_4$ was the first cuprate shown to exhibit *static* one-dimensional charge and spin stripes (in experiments performed at the High Flux Beam Reactor at Brookhaven National Laboratory by Tranquada, Axe, and coworkers). This observation provided an explanation for the previously known suppression of superconductivity in this material at 1/8 doping: *commensurate one-dimensional static charge and spin stripes compete with superconductivity*. Pressure is a convenient way to change the structure at constant chemical composition, and it is clear from our results that superconductivity is strongly enhanced by suppressing the structural phase transitions in $La_{1.48}Nd_{0.4}Sr_{0.12}CuO_4$. This observation suggests that the static charge and spin order is also eliminated by pressure. However, recent scanning tunneling microscope studies of other tetragonal cuprates in which the copper-oxygen planes are flat, as they are in the HTT structure of $La_{1.48}Nd_{0.4}Sr_{0.12}CuO_4$, have shown the presence of *two-dimensional* charge order. One way to reconcile these observations is to assume that the charge and spin order are one-dimensional in the LTT and LTO2 structures, but become two-dimensional in the HTT and LTO1 structures, with the latter situation more favorable toward superconductivity. This scenario would be consistent with the accumulating evidence for both one-dimensional and two-dimensional charge and spin order in the cuprates. Our results explicitly illustrate the impact of subtle changes of crystal structure upon superconductivity, and by implication upon charge and spin order, in these fascinating materials.

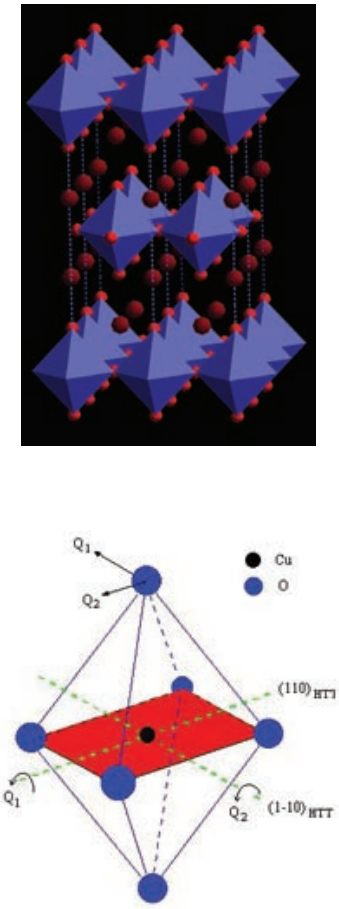

Figure 1. (Top) The crystal structure of La_2CuO_4, showing the two-dimensional layers of corner-sharing CuO_6 octahedra (blue), the oxygen atoms (light red), and the La atoms (dark red). (Bottom) A schematic view of a single CuO_6 octahedron illustrating the relative orientations of the (110) and (1-10) axes of the HTT structure, for which $|Q_1| = |Q_2| = 0$. The LTO1 structure is obtained by a single rotation of magnitude $|Q_1|$ or $|Q_2|$ about either of these axes (yielding one of the two orthorhombic twin structures). The LTO2 structure is obtained by rotations of magnitude $|Q_1|$ and $|Q_2|$ about both axes (where $|Q_1| \neq |Q_2|$). The LTT structure has rotations of equal magnitude about both axes, $|Q_1| = |Q_2| \neq 0$.

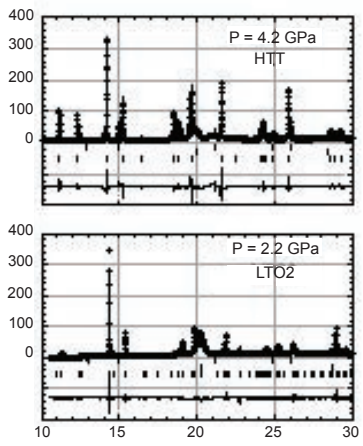

Figure 2. The x-ray powder diffraction patterns and results of General Structure Analysis Structure refinements for $La_{1.48}Nd_{0.4}Sr_{0.12}CuO_4$ at pressures of 4.2 GPa (top frame) or 2.2 GPa (bottom frame), at a temperature of 10 K. In the upper trace for each frame the crosses are the data, and the solid line is the calculated pattern; the lowest trace is the residual. The vertical tick marks are located at the positions of (top) NaCl included in the diamond anvil cell as an internal manometer, (middle) Fe due to the diamond anvil cell gasket, and (bottom) $La_{1.48}Nd_{0.4}Sr_{0.12}CuO_4$.

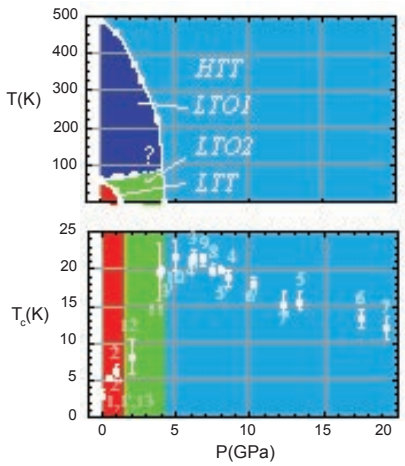

Figure 3. (Top) Pressure-temperature phase diagram for $La_{1.48}Nd_{0.4}Sr_{0.12}CuO_4$ determined by high-pressure x-ray powder diffraction. (Bottom) Superconducting transition temperature (T_c) versus pressure (P) for $La_{1.48}Nd_{0.4}Sr_{0.12}CuO_4$. In both frames the LTT phase region is shown in red, the LTO2 region in green, the LTO1 region in dark blue, and the HTT region in light blue.